

The mass-spectrometer technique used in the analysis of these samples was that described by some of us in connection with studies of lanthanum and cerium.<sup>3</sup> Aliquots of the samples were pipetted onto the filament of a surface ionization type source as a nitric acid solution. Gentle heating served to convert the nitrate to an adherent oxide coat. Emission of  $\text{Ga}^+$  ions began at approximately 650°C. No compound gallium ions were observed throughout the course of the experiment. The results obtained, along with the results of Sampson and Bleakney,<sup>4</sup> are given in Table I.

TABLE I. Abundance ratio of  $\text{Ga}^{69}/\text{Ga}^{71}$  from terrestrial and meteoritic sources.

Investigator	Source	$\text{Ga}^{69}/\text{Ga}^{71}$ ratio
Sampson and Bleakney <sup>4</sup>	Terrestrial	1.58
This work	Terrestrial	1.510
This work	Meteoritic	1.509

The samples were four times introduced, each for purposes of precision in the comparison of the isotopic ratios. Evaluation of errors shows that the isotopic ratios of the terrestrial and meteoritic samples are the same to within two-tenths of one percent. The absolute ratios given in Table I are the average values of 250 individual results. As a result of possible fractionations and discriminations they are good to three-fourths of one percent. It is apparent, however, that our value is not compatible with that of Sampson and Bleakney. This may be due to voltage effects in the Sampson and Bleakney measurements which were avoided in our observations.

<sup>1</sup> H. Brown and M. G. Inghram, Phys. Rev. 72, 347 (1947).

<sup>2</sup> H. Brown and E. Goldberg (work to be published).

<sup>3</sup> M. G. Inghram, R. J. Hayden, and D. C. Hess, Jr., Phys. Rev. 72, 967 (1947).

<sup>4</sup> M. B. Sampson and W. Bleakney, Phys. Rev. 50, 456 (1936).

### Long-Lived Tc Activities Produced by Deuteron Bombardment of Separated Mo Isotopes\*

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June 10, 1948

IN two earlier letters (April 30, 1948 and May 26, 1948) a preliminary report was given on a group of Tc activities (element 43) produced by irradiation of the separated Mo isotopes with ~16-Mev deuterons. It is now possible to report confirmatory evidence on the mass assignment of several of the known long-lived Tc activities, based on the results of bombardments of the various separated Mo isotopes with ca. 22-Mev deuterons from the Berkeley 60-inch cyclotron.

**4.2-day Tc.** This Tc activity<sup>1,2</sup> was produced in greatest yield from the deuteron bombardment of Mo enriched in  $\text{Mo}^{96}$  and decayed with a half-life of  $4.2 \pm 0.1$  days (Fig. 1). This result serves to confirm the assignment of the 4.2-day Tc to  $\text{Tc}^{96}$ . Gamma-rays of  $0.8 \pm 0.1$ -Mev energy as well as 17-18-kev x-rays were observed. Charged particles of 0.64-Mev energy<sup>2</sup> were not found, however.

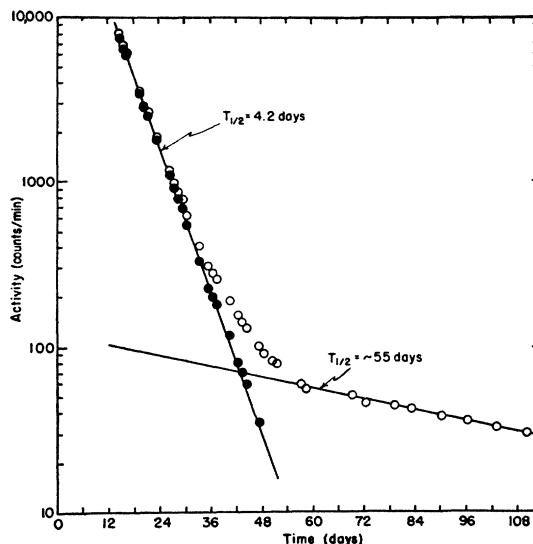


FIG. 1. Decay curve of the 4.2-day Tc activity produced by deuteron bombardment of Mo enriched in  $\text{Mo}^{96}$ .

**50-60-day Tc.** The previously studied 50-60-day  $\text{Tc}^{96}$  was produced in greatest yield from the deuteron bombardment of Mo enriched in  $\text{Mo}^{96}$ . The decay curve (Fig. 2A) gave an apparent half-life of 59 days. This value will be reduced slightly by the subtraction of a 90-day component which is almost certainly present. The radiations found associated with this period were a  $0.8 \pm 0.1$ -Mev  $\gamma$ -ray, a  $0.24 \pm 0.05$ -Mev  $\gamma$ -ray, and x-rays characteristic of the

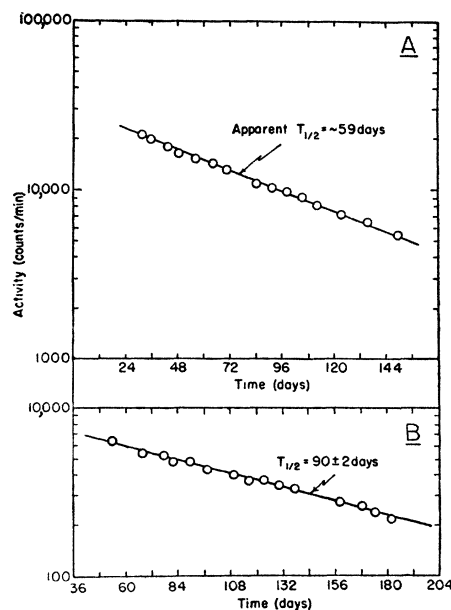


FIG. 2. A. Decay curve of the 50-60-day Tc activity produced by deuteron bombardment of Mo enriched in  $\text{Mo}^{96}$ . B. Decay curve of the 90-day Tc activity produced by deuteron bombardment of Mo enriched in  $\text{Mo}^{97}$ .

Mo-Tc region. The  $\gamma$ -ray of lower energy was twice as abundant as that of higher energy in agreement with Huber *et al.*<sup>4</sup> In addition, conversion electrons of  $\sim 180$ -keV energy were observed (visual range 40 mg Al/cm<sup>2</sup>). The predominantly greater yield of this 50–60-day Tc activity from Mo enriched in Mo<sup>96</sup> supports its assignment to Tc<sup>96</sup>.

**90-day Tc.** Our earlier work<sup>5</sup> on the approximately 90-day Tc had indicated its assignment to mass 97. It was to be expected, then, that a  $d,2n$  reaction would give the greatest yield of Tc<sup>97</sup> when carried out on Mo enriched in Mo<sup>97</sup>. This result was actually found and confirms the assignment of the 90-day Tc. Soft electrons of  $\sim 85$ -keV energy ( $\tau_{1/2} = 1.5$  mg Al/cm<sup>2</sup>), and 17–18-keV x-rays were again found associated with this period. The yield of any unconverted soft gamma-radiation ( $\sim 97$  keV) was so small that its presence was doubtful. The decay curve (Fig. 2B) gave a  $90 \pm 2$ -day half-life.

\* This document is based on work performed under Contract No. W-7405-eng.-26 for the Atomic Energy Project at Oak Ridge National Laboratory.

- <sup>1</sup> Ewing, Perry, and McCreary, *Phys. Rev.* **55**, 1136 (1939).  
<sup>2</sup> J. E. Edwards and M. L. Pool, *Phys. Rev.* **72**, 384 (1947).  
<sup>3</sup> E. N. Cacciapuoti and E. Segrè, *Phys. Rev.* **55**, 110 (1939).  
<sup>4</sup> Huber, Medicus, Preiswerk, and Steffen, *Phys. Rev.* **73**, 1211 (1948).  
<sup>5</sup> Motta, Boyd, and Brosi, *Phys. Rev.* **71**, 210 (1947).

### Increase in Vapor Pressure of Liquid Helium Due to He<sup>3</sup> in Solution\*

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June 16, 1948

WITH a thermal diffusion column<sup>1</sup> we have recently produced about 100 standard cm<sup>3</sup> of helium gas containing approximately 0.16 percent He<sup>3</sup>. This sample, therefore, contains about ten thousand times more He<sup>3</sup> than ordinary gas well-helium. It seemed, therefore, worth while to attempt to measure differences in the vapor pressure between this enriched gas and the normal well-helium, the latter being practically pure He<sup>4</sup>.

The apparatus consisted of two identical thin-walled copper vessels, each of volume approximately 0.16 cm<sup>3</sup>, the two being silver-soldered together. A pair of thin-walled stainless steel tubes ran from each vessel out of the cryostat to the respective legs of an oil manometer, the latter containing low vapor pressure diffusion pump oil (Octoil-S). The two levels in the manometer were read with a cathetometer, and with this apparatus vapor pressure differences of the order of  $10^{-3}$  mm Hg could be detected. The two copper vessels were surrounded by a bath of liquefied well-helium whose temperature could be controlled by pumping in the usual way. Corresponding absolute temperatures were computed from the vapor pressure of this bath using the Leiden 1937 tables.

The results of the measurement are shown in Fig. 1. As

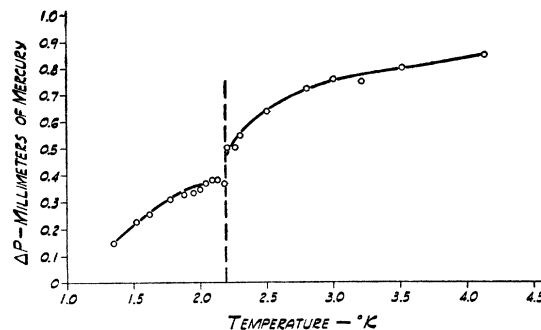


FIG. 1. The difference in vapor pressure ( $\Delta P$ ) between helium containing 0.16 percent He<sup>3</sup> and well-helium as a function of temperature. The vertical broken line passes through the  $\lambda$ -point.

was expected, the He<sup>3</sup>-enriched material showed a higher vapor pressure than the He<sup>4</sup> at all temperatures. In order to check the reliability of our apparatus we performed a blank experiment using well-helium in both copper vessels. Under these conditions, the vapor-pressure difference was less than the least count of our manometer ( $\sim 10^{-2}$  mm Hg) at all temperatures in the range shown in Fig. 1. In view of this it seems unlikely that the discontinuity at the  $\lambda$ -point shown in Fig. 1 is due to small-temperature inhomogeneities in the bath when it is in the He I region. Further, the results were quite reproducible.

The possibility that the  $\lambda$ -point discontinuity is due to a secondary effect connected with the creeping film is being investigated. One possibility is that the evaporated film on recondensing produces a downward heat flush in the liquid in the copper bulb disturbing the isotope distribution.<sup>2</sup>

If one assumes that this solution of He<sup>3</sup> in He<sup>4</sup> is ideal, that is, obeys Raoult's and Henry's laws, then it can be shown that the following relations hold:

$$\Delta P = C_L [(C_V/C_L) - 1] \pi_4, \quad (1)$$

$$\Delta P = C_L (\pi_3 - \pi_4). \quad (2)$$

Here the  $\pi$ 's represent the saturated vapor pressures of the two pure isotopes, and  $C_V$ ,  $C_L$  are, respectively, the He<sup>3</sup> to He<sup>4</sup> concentrations in the vapor and liquid. Previously values of  $C_V/C_L$  as a function of temperature in the He I region for very dilute solutions have been measured by us.<sup>3</sup> If we suppose that the much more concentrated (by about  $10^3$ ) solution we have here obeys the same relationship, then  $C_L$  as a function of temperature may be computed from this and the balance of mass and composition equations. This is tantamount to supposing Henry's law to be valid in this concentration range. Since now all quantities on the right of Eq. (1) are known,  $\Delta P$  may be computed as a function of temperature in the He I region. The results of this give only order-of-magnitude agreement, the computed values being around 40 percent lower than the measured values, but the calculation nevertheless suggests that, in the He I region, the solution is not far removed from an ideal classical solution. Similarly, from Eq. (2) a rough estimate may be made of the vapor-pressure curve<sup>4</sup> for He<sup>3</sup>. A plot of  $\log \pi_3$  versus  $1/T$  made from the above data gives a